

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

(Chapter II of the Patent Cooperation Treaty)

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference Cal 87701	FOR FURTHER ACTION	
See Form PCT/PEA/416		
International application No. PCT/EP2004/003673	International filing date (day/month/year) 06.04.2004	Priority date (day/month/year) 11.04.2003
International Patent Classification (IPC) or national classification and IPC C08F218/00, G02B1/04		
Applicant GREAT LAKES CHEMICAL (EUROPE) GMBH		

<p>1. This report is the international preliminary examination report, established by this International Preliminary Examining Authority under Article 35 and transmitted to the applicant according to Article 36.</p> <p>2. This REPORT consists of a total of 7 sheets, including this cover sheet.</p> <p>3. This report is also accompanied by ANNEXES, comprising:</p> <p>a. <input checked="" type="checkbox"/> (<i>sent to the applicant and to the International Bureau</i>) a total of 6 sheets, as follows:</p> <ul style="list-style-type: none"> <input type="checkbox"/> sheets of the description, claims and/or drawings which have been amended and are the basis of this report and/or sheets containing rectifications authorized by this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions). <input type="checkbox"/> sheets which supersede earlier sheets, but which this Authority considers contain an amendment that goes beyond the disclosure in the international application as filed, as indicated in item 4 of Box No. I and the Supplemental Box. <p>b. <input type="checkbox"/> (<i>sent to the International Bureau only</i>) a total of (indicate type and number of electronic carrier(s)) , containing a sequence listing and/or tables related thereto, in computer readable form only, as indicated in the Supplemental Box Relating to Sequence Listing (see Section 802 of the Administrative Instructions).</p>
<p>4. This report contains indications relating to the following items:</p> <ul style="list-style-type: none"> <input checked="" type="checkbox"/> Box No. I Basis of the opinion <input type="checkbox"/> Box No. II Priority <input type="checkbox"/> Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability <input type="checkbox"/> Box No. IV Lack of unity of invention <input checked="" type="checkbox"/> Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement <input type="checkbox"/> Box No. VI Certain documents cited <input checked="" type="checkbox"/> Box No. VII Certain defects in the international application <input type="checkbox"/> Box No. VIII Certain observations on the international application

Date of submission of the demand 09.11.2004	Date of completion of this report 17.03.2005
Name and mailing address of the international preliminary examining authority:  European Patent Office - P.B. 5818 Patentlaan 2 NL-2280 HV Rijswijk - Pays Bas Tel. +31 70 340 - 2040 Tx: 31 651 epo nl Fax: +31 70 340 - 3016	Authorized Officer Stabel, A Telephone No. +31 70 340-4158



**INTERNATIONAL PRELIMINARY REPORT
ON PATENTABILITY**

International application No.
PCT/EP2004/003673

Box No. I Basis of the report

1. With regard to the **language**, this report is based on the international application in the language in which it was filed, unless otherwise indicated under this item.
 - This report is based on translations from the original language into the following language, which is the language of a translation furnished for the purposes of:
 - international search (under Rules 12.3 and 23.1(b))
 - publication of the international application (under Rule 12.4)
 - international preliminary examination (under Rules 55.2 and/or 55.3)
2. With regard to the **elements*** of the international application, this report is based on (*replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report*):

Description, Pages

1-34 as originally filed

Claims, Numbers

1-21 filed with telefax on 09.11.2004

- a sequence listing and/or any related table(s) - see Supplemental Box Relating to Sequence Listing

3. The amendments have resulted in the cancellation of:
 - the description, pages
 - the claims, Nos.
 - the drawings, sheets/figs
 - the sequence listing (*specify*):
 - any table(s) related to sequence listing (*specify*):
4. This report has been established as if (some of) the amendments annexed to this report and listed below had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).
 - the description, pages
 - the claims, Nos.
 - the drawings, sheets/figs
 - the sequence listing (*specify*):
 - any table(s) related to sequence listing (*specify*):

* *If item 4 applies, some or all of these sheets may be marked "superseded."*

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Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes: Claims	1-21
	No: Claims	
Inventive step (IS)	Yes: Claims	
	No: Claims	1-21
Industrial applicability (IA)	Yes: Claims	1-21
	No: Claims	

2. Citations and explanations (Rule 70.7):

see separate sheet

Box No. VII Certain defects in the international application

The following defects in the form or contents of the international application have been noted:

see separate sheet

**INTERNATIONAL PRELIMINARY
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(SEPARATE SHEET)**

International application No.

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Reference is made to the following document/s/:

D1: EP-A-0 473 163 (ENICHEM SINTESI) 4 March 1992 (1992-03-04)
D3: WO 01/25301 A (GREAT LAKES CHEMICAL EUROP GMB ; GIROLDINI
WILLIAM (IT); CRISCI LUCIAN) 12 April 2001 (2001-04-12)

Re Item V

A Amendments

The amendments made are not complying with Article 19(2) / 34(2)(b) PCT and PCT International Preliminary Examination Guidelines (A20.21 which refers to the EPO Guidelines for examination); in particular the disclaimer "with the proviso that polyol C is not tris(hydroxy-ethyl) isocyanurate" is not allowable under Article 123(2) EPC (see also EPC Guidelines C VI 5.3.11 and decisions G01/03 and G02/03).

A positive formulation naming polyols from which can be selected, would be allowable under Article 19(2) / 34(2)(b) PCT.

B Novelty¹

D1 discloses a liquid polymerizable composition for organic glass used in ophthalmic lenses (page 2 line 7) comprising

- the product obtained by the transesterification of diallyl carbonate with a mixture comprising di-ethylene glycol and tris(hydroxyethyl) isocyanurate/or pentaerythritol and
- di-benzoyl peroxide as a free radical initiator in quantity of 0.08-0.33 mol/kg (2-8

¹ examined as if a positive formulation of the disclaimer had been used, namely allowing one of pentaerythrite, trimethylpropane, dipentaerythrite or di - trimethylol propane; otherwise D2 (US-A-5286816) remains novelty destroying for claims 1-6, 8-15 and 18-20

wt %) (see claim 8). The transesterification conditions (temperature range, catalyst, amount of catalyst, pressure and reaction time) are given in EP0302537 col. 2 lines 37-67 cited in D1 (page 3 lines 20-24). In example 4 (product (D)), the ratio of pentaerythritol to di-ethylene glycol is 25:75 wt. % and the ratio diallyl carbonate to diol/polyol is 5:1. The composition can further comprise an additional co-monomer (0-25 wt. %) of eg. acrylic, vinylic, allylic or methacrylic type; one example being vinyl acetate (claim 7).

The subject matter of claim 1 *differs* from the disclosure of D1 in that the concentration of radical polymerization initiator (BCHPC and/or di benzoyl peroxide) is limited to 0.03-0.1 mol/kg of final composition, while the range in D1 is limited to 0.08-0.33 mol/kg (claim 8). The man skilled in the art when not confronted with the task of solving a particular problem, but to reproduce the teaching of D1, would not seriously contemplate to work in the overlapping range² of 0.08-0.1 for the following reasons:

- (I) The range 0.08-0.33 disclosed in claim 8 is given for a polymerization initiator, while in claim 9 of D1 the range is limited to 0.12-0.24 mol/kg for the specific family of peroxide initiators to which both compounds of claim 1 of the present application belong; thus no overlap with the claimed range.
- (ii) All the examples given in D1 use 0.17 mol/kg which lies outside the range of present claim 1.

The subject matter of claims 1-21 is therefore considered novel.

C Inventive Step

C.1 The *effect* of the difference between the subject matter of claims 1, 2, 4-16, 18-21 and D1 is that the yellowness is smaller when less peroxide is used, while maintaining sufficient mechanical properties of the organic glass.

The remaining *problem* to be solved can be considered to be how to reduce yellowness, while maintaining the necessary mechanical properties of the organic glass.

It is obvious to the man skilled in the art starting from the teaching of D1 to reduce the

²cf. also T26/85

amount of peroxide initiator in order to solve the remaining technical problem mentioned above for the following reasons:

It has been disclosed in **D1** that the long known problem that organic glass produced according to the known techniques has various deficiencies mainly due to the excessive degree of colouring (generally yellow) which it acquires during the polymerization stage of the relative monomers, caused by the organic peroxides used as catalysts (page 2, lines 9-11), has been solved by adding a small quantity of a free aliphatic polyol to the liquid compositions to be polymerized, made up of or containing at least one bis(allyl carbonate) of diethylene glycol or at least one poly(allyl carbonate) of a di- or poly-hydroxy alcohol (page 2 lines 17-20).

D1 further discloses that the addition of a free aliphatic polyol does not interfere in any way with the other characteristics of the polymerized products, particularly its thermal and mechanical properties.

The man skilled in the art, starting from the teaching of **D1** including the examples thereof which show indeed a generally higher Yellow Index but also an increased Rockwell Hardness when compared to the examples of the present application, would when trying to reduce the yellowness without losing too much of the mechanical properties, simply by *routine experiments* gradually reduce the amount of peroxide initiator and thereby fall inevitably into the range claimed by the applicant. The man skilled in the art would expect that starting from the examples in **D1**, he would by reducing the peroxide concentration obtain an organic glass with a reduced yellowness and a decreased hardness, thereby getting indeed closer to the yellowness and hardness values of the present application.

C.2 **D2** discloses a liquid polymerizable composition for organic glass comprising

- the product obtained by the transesterification of diallyl carbonate with a mixture comprising di-ethylene glycol and tris(hydroxyethyl) isocyanurate
- and
- di-benzoyl peroxide as a free radical initiator in quantity of 0.04-0.25 mol/kg (1-6 wt %) (see claim 4). The composition can further comprise an additional co-monomer (0-25 wt. %) of eg. acrylic, vinylic, allylic or methacrylic type; one example being vinyl acetate (claim 7). The ratio of the sum of the diols and the diallyl carbonate is higher than 3/1 and the amount of polyol with respect to the diols is 10 - 40% (see claim 1).

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The transesterification conditions (temperature range, catalyst, amount of catalyst, pressure and reaction time) are given (claims 3, 5 and 6 and col. 2 lines 38-66).

The *difference* with the subject matter of claims 1-6, 8-15 and 18-20 is that the polyol is one of pentaerythrite, trimethylpropane, dipentaerythrite or di - trimethylol propane. The *effect* thereof is not recognisable to the international preliminary examination authority. The *problem* to be solved consist therefor in providing an alternative polyol in an organic glass composition further based on a transesterification product of di allyl carbonate and aliphatic diols.

The choice of one of the above mentioned polyols is merely one of several straightforward possibilities from which the skilled person would select, in accordance with circumstances, *without the exercise of inventive skill*, in order to solve the problem posed (cf. e.g. D1 and D3).

C.3 Objection due to lack of inventive step as set out under B.1 of the our written opinion dated 16.07.2004 against the subject matter of claim 17 is maintained.

The subject matter of claims 1-21 does not meet the requirements of Article 33(3) PCT with respect to inventive step.

D Industrial applicability

The subject matter of claims 1-21 meets the requirements of Article 33(4) PCT with respect to industrial applicability.

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(72)

CLAIMS

1. A liquid composition which can undergo radicalic polymerization into organic glass comprising the following components:
 - 5 1) the product obtained from the transesterification of diallyl carbonate (A) with a blend of one or more linear or branched aliphatic diols (B), containing from three to ten carbon atoms in the molecule, with a linear or branched aliphatic polyol (C), containing from four to
 - 10 twenty carbon atoms and from three to six hydroxyl groups in the molecule with the proviso that polyol (C) is not tris(hydroxy-ethyl) isocyanurate; said component 1) being present in an overall concentration ranging from 70 to 100% by weight with respect to the total weight of the
 - 15 mixture of components 1) and 2);
 - 2) one or more co-monomers of the acrylic, methacrylic, vinylic or allylic type and mixtures thereof, in an overall concentration ranging from 0 to 30% weight with respect to the total weight of the mixture of components
 - 20 1) and 2);
 - 3) a polymerization initiator or a mixture of two or more polymerization initiators, stable at room temperature, belonging to the group of peroxides and selected from di(4-t-butyl-cyclohexyl)peroxy-dicarbonate (BCHPC) and
 - 25 dibenzoyl-peroxide, in an overall concentration ranging

from 0.03 to 0.1 moles per 1 kg of final composition, with the proviso that when component 2) is absent, di(4-t-butyl-cyclohexyl)peroxy-dicarbonate (BCHPC) is used in combination with dibenzoyl-peroxide.

5 2. The composition according to claim 1, characterized in that the molar ratio A/(B+C) ranges from 2/1 to 5/1 and the amount of (C) in the mixture (B+C) is equal to or lower than 25% by weight on the total of said mixture (B+C).

10 3. The composition according to claim 1, characterized in that the molar ratio A/(B+C) ranges from 2.5/1 to 4/1 and the amount of (C) in the mixture (B+C) ranges from 5% weight to 20% by weight on the total weight of said mixture (B+C).

15 4. The composition according to claim 1, characterized in that the diols (B) are diethylene glycol, triethylene glycol, tetraethylene glycol, 1,4-butanediol, 1,6-hexanediol, 1,3-propanediol, neopentyl glycol, dipropylene glycol, 2,2,4-trimethyl-1,3-pentanediol, 1,4-cyclohexane dimethanol.

5. The composition according to claim 4, characterized in that the diols are diethylene glycol and neopentyl glycol.

6. The composition according to claim 1, characterized in that the polyols (C) are pentaerythrite, trimethyl

propane, dipentaerythrite, di-trimethylol propane, -
~~tris(hydroxyethyl) isocyanurate.~~

7. The composition according to claim 6, characterized in that the polyols (C) are pentaerythrite, trimethylol 5 propane.

8. The composition according to any of the previous claims, characterized in that component 1) is obtained starting from diallyl carbonate (A) and the mixture (B+C), operating under transesterification conditions, at 10 a temperature ranging from 80 to 160°C, in the presence of an alkaline-type catalyst, continuously eliminating the allyl alcohol which is formed as reaction by-product.

9. The composition according to claim 8, characterized in that the transesterification is carried out at a temperature ranging from 90 to 130°C and the alkaline catalyst is selected from hydroxides, carbonates and alcohohlates of alkaline metals, organic bases, basic ion exchange resins.

10. The composition according to claim 9, characterized 20 in that the catalyst is selected from sodium hydroxide, sodium carbonate, sodium methylate.

11. The composition according to any of the previous claims from 8 to 10, characterized in that the catalyst is used in an amount at least equal to 1 ppm (parts per 25 million by weight) with respect to the sum of the weights

of components (B+C).

12. The composition according to claim 11, characterized in that the catalyst is used in amounts ranging from 0.01% to 0.3% weight.

5 13. The composition according to any of the previous claims from 8 to 12, characterized in that the transesterification reaction is carried out at pressure values ranging from 60 mbar to 1030 mbar and for reaction times of between 0.5 and 20 hours.

10 14. The composition according to claim 13, characterized in that the transesterification reaction is carried out at pressure values ranging from 60 to 500 mbar.

15 15. The composition according to claim 13, characterized in that the transesterification reaction is carried out with reaction times ranging from 0.5 to 3 hours.

16. The composition according to claim 1, characterized in that component 2) is selected from methyl methacrylate, vinyl acetate, vinyl esters of versatic acids 9 and 10 known on the market as VeoVa 9 and VeoVa 10, triallyl 20 cyanurate, triallyl isocyanurate, diallyl maleate, diallyl fumarate, diallyl isophthalate, diallyl terephthalate and mixtures thereof.

17. The composition according to claim 1, characterized in that component 2) is present in an overall concentration ranging from 1 to 20% weight in the mixture of Com- 25

ponents 1) and 2).

18. The composition according to claim 1, characterized in that component 3) is selected from peroxides having a storage temperature not lower than +15°C.

5 19. The composition according to claim 1, characterized in that the peroxides are selected from dialkyl peroxy dicarbonates, diacyl peroxides and/or perketals.

20. The composition according to claim 19, characterized in that the dialkyl peroxy dicarbonates are di(4-t-butyl 10 cyclohexyl)peroxy dicarbonate and dimyristyl peroxy dicarbonate.

21. The composition according to claim 19, characterized in that the dialkyl peroxy dicarbonate is di(4-t-butyl cyclohexyl)peroxy dicarbonate (BCHPC).

15 22. The composition according to claim 19, characterized in that the diacyl peroxides are dibenzoyl peroxide, didecanoyl peroxide and dilauroyl peroxide.

23. The composition according to claim 19, characterized in that the diacyl peroxide is dibenzoyl peroxide.

20 24. The composition according to claim 19, characterized in that the perketals are 1,1-di(t-butyl peroxy) cyclohexane, 1,1-di(t-butyl peroxy)3,3,5-trimethyl cyclohexane, 1,1-di(t-amyl peroxy) cyclohexane.

25. The composition according to claim 19, characterized in that the perketal is 1,1-di(t-amyl peroxy) cyclohex-

ane.

26. The composition according to claim 1, characterized in that, when component 2) is absent, the diaetyl peroxides and/or perketals are used in combination with dialkyl peroxy dicarbonates.

18. 27. The composition according to any of the previous claims, characterized in that it contains one or more conventional additives, such as antioxidants, light stabilizers, lubricants, dyes, pigments, UV-absorbers, IR-absorbers, and similar, in a total amount in any case not higher than 1 part by weight for every 100 parts by weight of the compositions.

19. 28. The composition according to any of the previous claims, characterized in that it is transformed into the relevant organic glass by operating at a temperature ranging from 30 to 120°C, with polymerization times varying from 1 hour to 100 hours.

20. 29. Organic glass obtained by the polymerization of a composition according to any of the previous claims.

20 21. 30. Ophthalmic lenses, sun glasses, protective shields, display windows, manifolds and solar and photovoltaic panels, substrates for optical disks, display panels and video-terminals which can be obtained by the processing of the organic glass according claim 20. 29.